Supplemental Material: Quantum Dial for High-Harmonic Generation

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I. Tight-Binding Model

Using the tight-binding model, we have obtained the Hamiltonian identical to that presented in work [1] as follows

$$H_{11} = 2t_0 \left[\cos(k_x a) + 2\cos(\tfrac{ak_x}{2})\cos(\tfrac{\sqrt{3}k_y}{2})\right] + \epsilon_{11}, \quad \text{(S1)}$$

$$H_{12} = 2it_1 \sin(ak_x) + 2it_1 \sin(\tfrac{ak_x}{2})\cos\tfrac{\sqrt{3}ak_y}{2} - 2\sqrt{3}t_2 \sin(\tfrac{ak_x}{2})\sin(\tfrac{\sqrt{3}ak_y}{2}), \quad \text{(S2)}$$

$$H_{13} = 2t_2 \cos(ak_x) + 2i\sqrt{3}t_1 \cos(\tfrac{ak_x}{2})\sin(\tfrac{\sqrt{3}ak_y}{2}) - 2t_2 \cos(\tfrac{ak_x}{2})\cos(\tfrac{\sqrt{3}ak_y}{2}), \quad \text{(S3)}$$

$$H_{22} = 2t_{11}\cos(ak_x) + \cos(\tfrac{ak_x}{2})\cos(\tfrac{a\sqrt{3}k_y}{2})(t_{11} + 3t_{22}) + \epsilon_{22}, \quad \text{(S4)}$$

$$H_{23} = 2it_{12}\left[\sin(ak_x) - 2\sin(\tfrac{ak_x}{2})\cos(\tfrac{\sqrt{3}ak_y}{2})\right] - \sqrt{3}(t_{11} - t_{22})\sin(\tfrac{ak_x}{2})\sin(\tfrac{\sqrt{3}ak_y}{2}) \quad \text{(S5)}$$

$$H_{33} = 2t_{22}\cos(k_x a) + (3t_{11} + t_{22})\cos(\tfrac{k_x a}{2})\cos(\tfrac{a\sqrt{3}k_y}{2}) + \epsilon_{33}, \quad \text{(S6)}$$

where $H_{nm} = H^*_{mn}$. Fitting parameters using the generalized-gradient approximation are presented in Table. I [1].

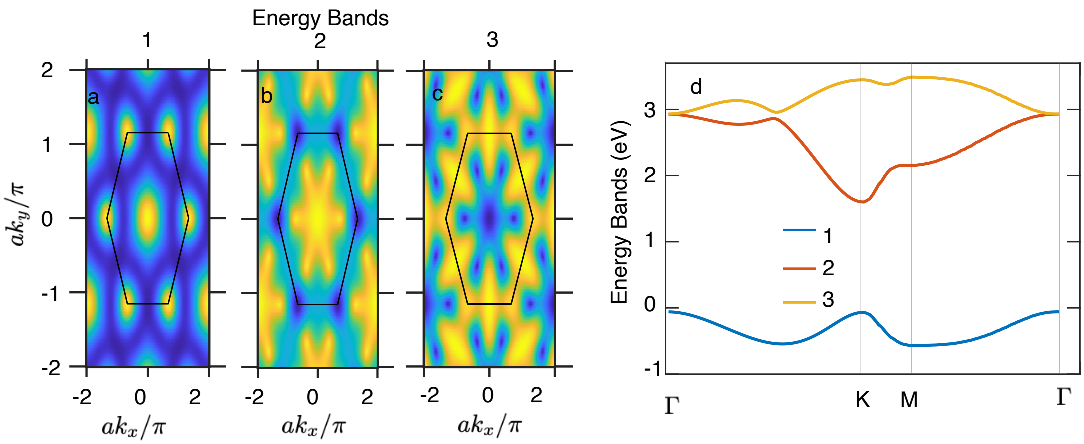

FIG. S1. The bands with energy from low to high are presented in panels a-c, respectively. The hexagonal black lines indicate the Brillouin zone. Panel d shows the energies of 3 bands along the high symmetry points Γ-K-M-Γ. The definitions of the symmetry points can be found in Section

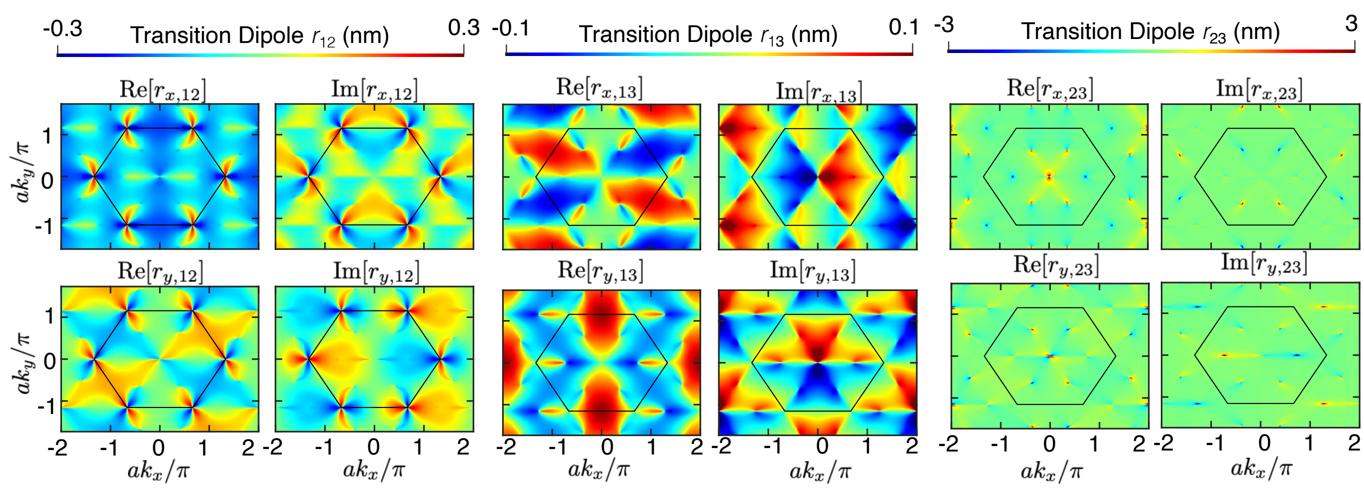

FIG. S2. The transition dipoles r_{12}, r_{13}, r_{23} are presented from left to right. The dipole components along x and y dimensions are shown on the top and bottom panels, respectively. Solid lines represent the Brillouin zone boundaries.

The visualizations of the bands (i.e. Eigen energies) obtained from Eqs. (S1-S6)are presented in Fig. S1. The corresponding transition dipoles are presented in Fig.S2.

	a(Å)	ϵ_1	ϵ_2	t_0	t_1	t_2	t_{11}	t_{12}	t_{22}
MoS_2	3.190	1.046	2.104	-0.184	0.401	0.507	0.218	0.338	0.057
WS_2	3.191	1.130	2.275	-0.206	0.567	0.536	0.286	0.384	-0.061
$MoSe_2$	3.326	0.919	2.065	-0.188	0.317	0.456	0.211	0.290	0.130
WSe_2	3.325	0.943	2.179	-0.207	0.457	0.486	0.263	0.329	0.034
$MoTe_2$	3.557	0.605	1.972	-0.169	0.228	0.390	0.207	0.239	0.252
WTe_2	3.560	0.606	2.102	-0.175	0.342	0.410	0.233	0.270	0.190

TABLE I. Fitted parameters of the three-band tight-binding model of monolayer MX_2. The lattice constant is denoted by a. The energy parameters are in units of eV. Parameters presented in this table are obtained from work [1]

A. Brillouin Zone Definition

Since the MoS_2 has a honeycomb lattice structure, in real space, the basis vectors can be chosen as

$$a_1 = a\hat{i} = (a, 0, 0), \quad a_2 = -a/2\hat{i} + \sqrt{3}a/2\hat{j} = (-a/2, \sqrt{3}a/2, 0), \quad a_3 = 1\hat{k} = (0, 0, 1) \tag{S7}$$

where $\hat{i}, \hat{j}, \hat{k}$ are unit vectors along x, y, z dimensions, respectively. Consequently, in the reciprocal space, we have basis vectors

$$b_1 = \frac{2\pi}{V} a_2 \times a_3 = \frac{4\pi}{\sqrt{3}a}(\frac{\sqrt{3}}{2}, \frac{1}{2}, 0), \quad b_2 = \frac{2\pi}{V} a_3 \times a_1 = \frac{4\pi}{\sqrt{3}a}(0, 1, 0), \quad b_3 = \frac{2\pi}{V} a_1 \times a_2 = \frac{4\pi}{\sqrt{3}a}(0, 0, \frac{\sqrt{3}}{2}), \tag{S8}$$

where $V = a_1 \cdot (a_2 \times a_3) = \sqrt{3}a^2/2$. Since we focus on the 2D material, the relevant reciprocal space is also 2D (i.e. only b vectors within the xy plane are relevant). As a result, in the following discussions, the z dimension is omitted. One can choose different a vectors in the real space, which will lead to different d vectors in the reciprocal space. The first Brillouin zone is formed by surfaces (3D lattice) or lines (2D lattice) that are perpendicular to the b vectors at half of the length (see Fig. S3**a**). The center of the Brillouin zone is $\Gamma = (0, 0)$. The corner of the Brillouin zone is denoted as by $K = (4\pi/3a, 0)$. The center of an edge is denoted by $M = (\pi/a, \pi/\sqrt{3}a)$ [2].

To obtain the physical variables such as the nonlinear current j, integration within the first Brillouin zone in the reciprocal space is required. For a hexagonal Brillouin zone as shown in Fig.S3**a**, one can use the Monkhorst Pack grid [3], which is a regime enclosed by the rhombus indicated by the dashed lines in Fig.S3**b** [4]. This rhombus regime with an edge length of $4\pi/(\sqrt{3}a)$ has the same area as a single hexagon. The same color indicates the equivalent areas. For the Monkhorst Pack grid, instead of the orthogonal coordinates $\hat{k}_x$, $\hat{k}_y$ mesh in the reciprocal space, we choose bases vectors along $\hat{b}_1$ and $\hat{b}_1''$ (shown in S3**a**) that are parallel to the rhombus edges as basis vectors. Thus, we have

$$v_k = \boldsymbol{B} v_b, \quad \boldsymbol{B} = \begin{pmatrix} \sqrt{3}/2 & -\sqrt{3}/2 \\ 1/2 & 1/2 \end{pmatrix}, \quad \boldsymbol{B^{-1}} = \begin{pmatrix} 1/\sqrt{3} & 1 \\ -1/\sqrt{3} & 1 \end{pmatrix} \tag{S9}$$

where v_b represents vector written by Monkhorst grid and v_k is the one represented by standard k_x, k_y orthogonal grid. For example, the corner of the Monkhorst grid is $v_b = (2\pi/(a\sqrt{3}), 2\pi/(a\sqrt{3}))$, which correspond to $k = (0, 2\pi/(a\sqrt{3})) = \boldsymbol{B} v_b$ after transformation. Consequently, the derivatives transform as follows

$$\partial_{k_x} f = \partial_{b_1} f \frac{\partial b_1}{\partial k_x} + \partial_{b_1''} f \frac{\partial b_1''}{\partial k_x} \tag{S10}$$

$$\partial_{k_y} f = \partial_{b_1} f \frac{\partial b_1}{\partial k_y} + \partial_{b_1''} f \frac{\partial b_1''}{\partial k_y}. \tag{S11}$$

As a result, we have

$$\nabla_{\boldsymbol{k}} = (B^{-1})^T \nabla_{\boldsymbol{b}}. \tag{S12}$$

Thus, the integration over the entire Brillouin zone is

$$\frac{1}{V_{BZ}} \iint f(k_x, k_y) dk_x dk_y = \frac{1}{V_{BZ}} \iint f(b_x, b_y) \det(\boldsymbol{B}) db_x db_y, \tag{S13}$$

where $V_{BZ} = 8\pi^2/(\sqrt{3}a^2)$ is the area of the Brillouin zone. Since the angle between the basis vectors (adjacent edges of the rhobus) in the Monkhorst grid is $\pi/3$ degrees, we should have the relation $Det(B) = \sin(\pi/3)$ to conserve the area in Eq.(S13).

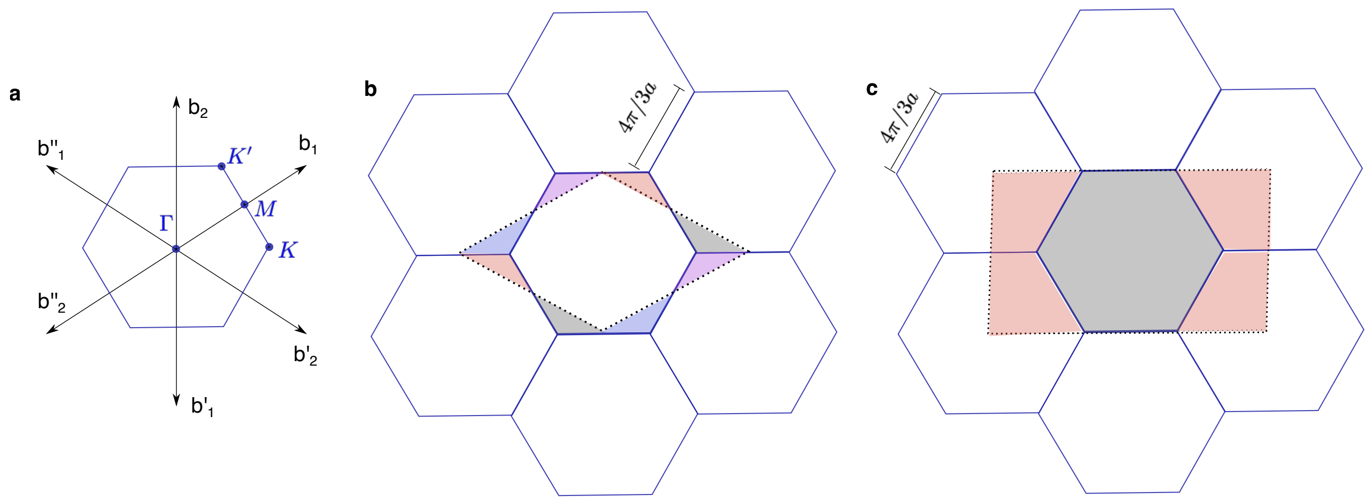

FIG. S3. Panel **a** shows the definition of the Brillouin zone and high symmetry points. Panel **b** indicates the definition of the Monkhorst Pack i.e. the rhombus regime enclosed by the dashed lines. This rhombus regime has the same area as a single hexagon. Owing to the periodicity, the equivalent areas are indicated by the same color. Panel **c** illustrates a rectangular mesh space that contains 2 Brillouin zones.

One may think a square regime in the Brillouin zone with standard vertical k-space meshes would be equivalent, which is shown in Fig.S3**c** marked by a dashed square box. This regime is periodic and contains 2 Brillouin zones, where different colors mark different Brillouin zones. Unfortunately, this will very likely lead to numerical errors because most transitions typically occur around the K and K' points, but now these points are at the calculation boundaries. For the Monkhorst grid, the K and K' points are contained well within the calculation domain.

Now, we can generate the current with both the density matrix elements' time evolution and the tight-binding model's Hamiltonian. Since transition dipole moment $r_{nm,n\neq m} = \eta_{nm} = \eta^*_{mn}$, for an N level system (here we have chosen $N = 3$), the number of independent transition dipole moment is $N \times (N-1)/2$.

II. Define the Hamiltonian

We start with the Spin-Boson model as follows [5]

$$\bar{H} = -\frac{\mathcal{E}(\boldsymbol{K}_t,t)}{2}\sigma_z + \frac{1}{2}\hbar\Omega(\boldsymbol{K}_t,t)\sigma_x + \sum_q \hbar\omega_q b_q^\dagger b_q + \sigma_z \sum_q g_q \left(b_q + b_q^\dagger\right). \tag{S14}$$

Here, $\mathbf{E}(t)$ is the laser electric field, the vector potential is defined by $-\partial_t \mathbf{A} = \mathbf{E}$, the Hamiltonian is represented in the canonical momentum $\boldsymbol{K}$ frame and we denote $\mathbf{K}_t = \mathbf{K} + e\mathbf{A}(t)/\hbar$, where $\mathbf{K}$ is defined in the shifted Brillouin zone $\overline{\mathrm{BZ}}$. Further, $\Omega(\boldsymbol{K}_t,t) = (2e/\hbar)\mathbf{d}(\boldsymbol{K}_t,t)\mathbf{E}(t)$ is a generalized Rabi frequency, $e > 0$ is the elementary charge and $\hbar$ is the reduced Planck constant; $\mathbf{d}(\boldsymbol{K}_t,t)$ and $\mathcal{E}(\boldsymbol{K}_t,t)$ represent transition dipole and bandgap between conduction $|1\rangle$ and valence $|0\rangle$ band, respectively. Note that we have chosen a gauge such that the Rabi frequency is always a real number. The time dependence of these quantities arises from the shifted Brillouin zone. The Pauli matrices are denoted by σ_j $(j = x,y,z)$. Finally, ω_q, $\hat{b}_q^\dagger$, $\hat{b}_q$ and g_q are the harmonic oscillator frequency, creation, and annihilation operators, and the coupling coefficient of a mode with momentum $\mathbf{q}$, respectively. Following the approaches of our previous work [5] and performing the polar transform $H = UH_0H^\dagger$ to the original Hamiltonian H_0 by

$$U = \exp\left[\sigma_z \sum_q \left(\frac{g_q^*}{\hbar\omega_q} b_q^\dagger - \frac{g_q}{\hbar\omega_q} b_q\right)\right] = \begin{bmatrix} D^\dagger & 0 \\ 0 & D \end{bmatrix}, \quad D = \exp\left[-\sum_q \left(\frac{g_q^*}{\hbar\omega_q} b_q^\dagger - \frac{g_q}{\hbar\omega_q} b_q\right)\right], \tag{S15}$$

we obtain

$$H = H_B + H_I. \tag{S16}$$

In particular

$$H_B = \sum_q \hbar\omega_q b_q^\dagger b_q, \quad H_I = -\frac{\mathcal{E}}{2}\sigma_z + \frac{1}{2}\hbar\Omega(t)\left(\sigma_+ {D^\dagger}^2 + \sigma_- D^2\right), \tag{S17}$$

where $\sigma_+ = (\sigma_x + i\sigma_y)/2$ and $\sigma_- = (\sigma_x - i\sigma_y)/2$. By going to the interaction picture via the transformation

$$\hat{\rho} = \exp(-\frac{iH_B t}{\hbar})\hat{\rho}_I \exp(\frac{iH_B t}{\hbar}), \tag{S18}$$

we obtain the Liouville–Von Neumann equation as

$$i\hbar\partial_t \rho_I = [H_I(t), \rho_I]. \tag{S19}$$

Here we have used

$$H_I(t) = \exp(iH_B t/\hbar) H_I \exp(-iH_B t/\hbar) = \begin{bmatrix} -\mathcal{E} & D^\dagger(t)^2\hbar\Omega(t) \\ D(t)^2\hbar\Omega(t) & \mathcal{E} \end{bmatrix}, \tag{S20}$$

where $D(t) = \exp\left\{-\sum_q\left[\frac{g_q^*}{\hbar\omega_q} b_q^\dagger(t) - \frac{g_q}{\hbar\omega_q} b_q(t)\right]\right\}$, and $b_q(t) = b_q \exp(-i\omega_q t)$.

A. Initial conditions

Since H in Eq.(S16) is obtained by polar transformation $H = UH_0H^\dagger$ [5], the initial condition of the density matrix in the polar frame has to be obtained accordingly. Because $H_0 = i\hbar\partial_t|\Psi_0\rangle$ and $UH_0U^\dagger U|\Psi_0\rangle = H_p|\Psi_p\rangle$, the wave function transforms as $|\Psi_p\rangle = U|\Psi_0\rangle$ and the density matrix in the polar frame is written as

$$\hat{\rho}_p = U\hat{\rho}_0 U^\dagger. \tag{S21}$$

We start with the overall initial condition as an outer product $\otimes$ of the two-level system and the environment B stems from the squeezed vacuum as follows

$$\hat{\rho}_0(t=0) = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \otimes B, \quad B = |\xi\rangle\langle\xi| \tag{S22}$$

where $\beta = 1/(k_B T)$,k_B is the Boltzmann constant and T is the temperature and $|\xi\rangle$ is the squeezed vacuum. The multi-mode squeezed vacuum in plane wave bases is generated from the vacuum photon number state $|0\rangle$ as follows [6–9]

$$|\xi\rangle = \hat{S}(\xi)|0\rangle, \quad \hat{S}(\xi) = \exp\left[\frac{1}{2}\sum_{i,j}\left(\xi_{ij}^* a_i a_j - \xi_{ij} a_i^\dagger a_j^\dagger\right)\right]. \tag{S23}$$

The exponential term in the multi-mode squeezed parameter originates from the fact that the brightly squeezed vacuum is typically generated through a second-order nonlinear process, such as optical parametric amplification. We define the pump pulse as a classical electric field as

$$E_p = E_{0p}\exp(-t^2/\tau^2)\exp(i\omega_p t) + c.c = \frac{E_{0p}\tau}{\sqrt{2}}\int \exp[-(\omega-\omega_p)^2\tau^2/4]\exp(-i\omega t)d\omega + c.c, \tag{S24}$$

where ω_p is the carrier frequency. The energy conservation requires $\omega_p = \omega_i + \omega_j$. When $i = j$, it is related to the degenerate case where $\omega_i = \omega_j = \omega_p/2$. For $i \neq j$ $\omega_i \neq \omega_j \neq \omega_p/2$. Since the electric field operator of a given frequency ω_i is written as $\hat{E}_i = iE_0 a_i^\dagger \exp(i\omega_i) - iE_0 a_i \exp(-i\omega_i)$, we can write down the interaction Hamiltonian of the BSV as the product of the pump pulse and the polarization P as [7]

$$H_I = E_p \cdot P = V\epsilon_0\chi^{(2)}\sum_{i,j} E_p(t)E_i(t)E_j(t) \tag{S25}$$

$$\propto \chi^{(2)}E_{0p}\hbar\sum_{i,j}\sqrt{\omega_i\omega_j}\exp\left(-\frac{t^2}{\tau^2}\right)a_i^\dagger a_j^\dagger \exp\left[i(\omega_i+\omega_j-\omega_p)\right] + H.C., \tag{S26}$$

where V is the volume of the system, $H.C.$ represents the hermitian conjugate. We used the conservation of energy, i.e., the relevant frequency we want to focus on is the sum frequency case when $\omega_p = \omega_i + \omega_j$, and all the rest will not be phase-matched during the optical parametric process. As a result, the cross terms in Eq.(S25) are not relevant to us. Since the evolution of a wavefunction can be written as

$$i\hbar\partial_t |\Phi(t)\rangle = H_I |\Phi(t)\rangle . \tag{S27}$$

Substituting the H_I in Eq.(S26) into Eq.(S27) and use the relation in Eq.(S24), we obtain

$$\begin{aligned}|\Phi(t)\rangle &= \exp\left(-i\int H_I dt/\hbar\right)|\Phi(0)\rangle \propto \exp\left\{-i\chi^{(2)}E_{0p}\tau\sum_{i,j}\sqrt{\pi\omega_i\omega_j}\exp\left[-\frac{(\omega_p-\omega_i-\omega_j)\tau^2}{4}\right]a_i^\dagger a_j^\dagger + H.C.\right\}|\Phi(0)\rangle \\ &= \exp\left[\frac{1}{2}\sum_{i,j}\left(\xi_{ij}^* a_i a_j - \xi_{ij}a_i^\dagger a_j^\dagger\right)\right]|\Phi(0)\rangle . \end{aligned}\tag{S28}$$

We can now see that the operator in Eq. (S28) is exactly the commonly used multi-mode squeeze operator in Eq. (S23). We can write ξ_{ij} into a matrix form as $\boldsymbol{\xi}$, and it is clear that $\boldsymbol{\xi}$ is a complex symmetric matrix $\xi_{ij} = \xi_{ji}$. As a result, by denoting $\hat{a} = [a_1, a_2, ...a_N]$ and $\widetilde{\hat{a}}$ as transpose of $\hat{a}$, one can write the general squeezing operator in Eq.(S23) as

$$\hat{S} \equiv \exp\left(\frac{\widetilde{\hat{a}}\boldsymbol{\xi}^\dagger\hat{a}}{2} - \frac{\widetilde{\hat{a}}^\dagger\boldsymbol{\xi}\hat{a}^\dagger}{2}\right) \tag{S29}$$

By using the polar decomposition and writing $\boldsymbol{\xi} = \boldsymbol{r}\exp(i\boldsymbol{\theta})$, where the $\boldsymbol{r}$ and $\boldsymbol{\theta}$ are real and symmetric matrices. Besides, $\boldsymbol{\xi}\boldsymbol{\xi}^\dagger = \boldsymbol{r}^2$, $\boldsymbol{\xi}^\dagger\boldsymbol{\xi} = \exp(-i\boldsymbol{\theta})\boldsymbol{r}^2\exp(i\boldsymbol{\theta})$, and $\boldsymbol{r}^\dagger = \widetilde{\boldsymbol{r}}$. Using the baker-campbell-hausdorff formula $\exp(A)B\exp(-A) = B + [A,B] + \ldots \overbrace{[A[A,..[A},B]\ldots]]^{n}/n!$, we obtain [9, 10]

$$\hat{S}^{-1}a_p^\dagger\hat{S} = \sum_j a_j^\dagger(\cosh\widetilde{\boldsymbol{r}})_{jp} - a_j[\sinh(\widetilde{\boldsymbol{r}})\exp(-i\widetilde{\boldsymbol{\theta}})]_{jp} \tag{S30}$$

$$\hat{S}^{-1}a_p\hat{S} = \sum_j a_j(\cosh\boldsymbol{r})_{jp} - a_j^\dagger[\exp(i\boldsymbol{\theta})\sinh(\boldsymbol{r})]_{jp}. \tag{S31}$$

We denote the vacuum as

$$|0\rangle = |0_{q_1}, 0_{q_2}\cdots\rangle , \tag{S32}$$

which consists of the vacuum with bosons with different momentum q_i, $\quad i \in \{0, 1, 2, \cdots\}$. Here $\xi = re^{i\theta}$, r is known as the squeeze parameter and $0 \le r < \infty$ and $0 \le \theta \le 2\pi$. One can see that the operator $\hat{S}(\xi)$ is related to the two-photon process. Since $\hat{S}^{-1}(\xi) = \hat{S}^\dagger(\xi) = \hat{S}(-\xi)$, we know that the squeezed state is normalized i.e. $\langle\xi|\xi\rangle = 1$. Consequently, we have

$$\mathrm{Tr}\left[|\xi\rangle\langle\xi|\right] = \mathrm{Tr}\left[\hat{S}(\xi)|0\rangle\langle 0|\hat{S}^\dagger(\xi)\right] = \mathrm{Tr}\left[|0\rangle\langle 0|\right] = \sum_{n=0}\langle n|0\rangle\langle 0|n\rangle = 1. \tag{S33}$$

According to Eqs(S21,S22), we obtain the initial condition in the polar frame as

$$\hat{\rho}_p(t=0) = \begin{bmatrix} D^\dagger|\xi\rangle\langle\xi|D & 0 \\ 0 & 0\end{bmatrix}. \tag{S34}$$

To obtain the initial condition in the interaction picture, we perform the same transformation as in Eq. (S18) and obtain

$$\hat{\rho}_I(t=0) = \begin{bmatrix} \exp\left(\frac{iH_Bt}{\hbar}\right)D^\dagger|\xi\rangle\langle\xi|D\exp\left(-\frac{iH_Bt}{\hbar}\right) & 0 \\ 0 & 0\end{bmatrix}\Big|_{t=0} \tag{S35}$$

$$= \begin{bmatrix} D^\dagger|\xi\rangle\langle\xi|D & 0 \\ 0 & 0\end{bmatrix}. \tag{S36}$$

Note that the initial conditions for the squeezed vacuum and the thermal heat bath are different. The differences stem from the fact that the thermal heat bath lasts from $t = -\infty$ to ∞, whereas the squeezed vacuum appears as a pulse disturbance to the system. For the thermal heat bath, before any external excitation, the thermal heat bath already exists and is in equilibrium with the two-level system in the original H_0 frame, such that one can not write the density matrix in a decoupled outer product form of the two-level system and the environment as in Eq.(S22). However, after the polar transform, the environment and the two-level system are decoupled in the polar frame, judging from the H_p. Thus, the initial condition for the heat bath can be written as a direct outer product in the polar frame, as shown in Eq. (S39). On the other hand, the squeezed vacuum is triggered by an external excitation. Thus, before this excitation, nothing happens to the H_0 system, and the initial condition of the squeezed vacuum is written as Eq.(S22) i.e., it is in a decoupled form in the H_0 frame. To conclude, the initial condition in the interaction frame can be written as

$$\hat{\rho}_I(t=0) = \begin{bmatrix} \mathcal{B} & 0 \\ 0 & 0 \end{bmatrix}. \tag{S37}$$

In the following content, we denote

$$\text{Squeezed vacuum,} \quad \mathcal{B} = D^\dagger \left|\xi\right\rangle \left\langle\xi\right| D, \tag{S38}$$

$$\text{Thermal heat bath,} \quad \mathcal{B} = \exp\,(-\sum_q \hbar\omega_q b_q^\dagger b_q)/\mathrm{Tr_B}[\exp\,(-\sum_q \hbar\omega_q b_q^\dagger b_q)]. \tag{S39}$$

III. Calculate velocity operator

Since the high harmonic emission spectrum is obtained from the current operator $\hat{j} = -e\hat{v}$, the task reduces to obtaining the expectation value of the current $-e\left\langle\Psi\right|\hat{v}\left|\Psi\right\rangle$, where Ψ is the wave function. Since the Bloch basis and the photon number states are a complete set of bases i.e. $\sum_{m,n}\left|u_m\right\rangle\left|n\right\rangle\left\langle n\right|\left\langle u_m\right| = \mathbb{1}$, we have

$$\begin{aligned}\left\langle\Psi\right|\hat{v}\left|\Psi\right\rangle &= \sum_{m,n,k,l}\left\langle\Psi|u_k\right\rangle\left|l\right\rangle\left\langle l\right|\left\langle u_k\right|\hat{v}\left|u_m\right\rangle\left|n\right\rangle\left\langle n\right|\left\langle u_m|\Psi\right\rangle \\ &= \sum_{m,n,k,l}\left\langle n\right|\hat{\rho}_{mk}\left|l\right\rangle\left\langle l\right|v_{km}\left|n\right\rangle \\ &= \sum_{m,n,k}\left\langle n\right|\hat{\rho}_{mk}v_{km}\left|n\right\rangle,\end{aligned} \tag{S40}$$

where $v_{nm} = \left\langle u_n\right|\hat{v}\left|u_m\right\rangle$. Note that in the original frame, the $\hat{v}$ only operates on the electronic system, not the environment, i.e. the photon number states. However, since the polar transform in Eq.(S15) couples the environment and the two-band system, the $\hat{v}_I$ operator also operates on the environment. Similarly to steps in obtaining Eq. (S40), we can have in a different frame that

$$\left\langle\Psi_I\right|\hat{v}_I\left|\Psi_I\right\rangle = \sum_{m,n,k}\left\langle n\right|\hat{\rho}_{I,mk}v_{I,km}\left|n\right\rangle. \tag{S41}$$

We know that the choice of coordinates should not influence the value of the observable. This leads to $\left\langle\Psi_I\right|\hat{v}_I\left|\Psi_I\right\rangle = \left\langle\Psi\right|\hat{v}\left|\Psi\right\rangle$, which is equivalent to

$$\sum_{m,n,k}\left\langle n\right|\hat{\rho}_{mk}v_{km}\left|n\right\rangle = \sum_{m,n,k}\left\langle n\right|\hat{\rho}_{I,mk}v_{I,km}\left|n\right\rangle = \sum_n\left\langle n\right|\hat{\rho}_{I,11}v_{I,11} + \hat{\rho}_{I,22}v_{I,22} + \hat{\rho}_{I,12}v_{I,21} + \hat{\rho}_{I,21}v_{I,12}\left|n\right\rangle, \tag{S42}$$

where the last equality is the form specifically for a two-band system. Equation (S42) suggests that we need to multiply the density matrix element and the velocity element consistently in the same frame and then do the trace over the photon number states (for example, the heat bath) directly without any coefficients. In particular, the velocity operator in the interaction frame can be written as

$$\begin{aligned}\hat{v}_I &= \exp\left(\frac{iH_Bt}{\hbar}\right)U\hat{v}U^\dagger\exp\left(\frac{-iH_Bt}{\hbar}\right) = \exp\left(\frac{iH_Bt}{\hbar}\right)U\begin{bmatrix} v_{11} & v_{12} \\ v_{21} & v_{22}\end{bmatrix}U^\dagger\exp\left(\frac{-iH_Bt}{\hbar}\right) \\ &= \begin{bmatrix} v_{11} & v_{12}\exp\left(\frac{iH_Bt}{\hbar}\right)D^{\dagger\,2}\exp\left(\frac{-iH_Bt}{\hbar}\right) \\ v_{21}\exp\left(\frac{iH_Bt}{\hbar}\right)D^2\exp\left(\frac{-iH_Bt}{\hbar}\right) & v_{22}\end{bmatrix} \\ &= \begin{bmatrix} v_{11} & v_{12}D(t)^{\dagger\,2} \\ v_{21}D(t)^2 & v_{22}\end{bmatrix}.\end{aligned} \tag{S43}$$

One should be aware that the operator $D(t)$ commutes with the v_{ij} with $i, j \in \{1, 2\}$. We choose to calculate the harmonic emission in the polar frame. The resulting expectation value is

$$\langle \Psi_I | \hat{v}_I | \Psi_I \rangle = v_{11} \sum_n \langle n | \hat{\rho}_{I,11} | n \rangle + v_{22} \sum_n \langle n | \hat{\rho}_{I,22} | n \rangle + v_{12} \sum_n \langle n | \hat{\rho}_{I,21} D(t)^{\dagger\, 2} | n \rangle + v_{21} \sum_n \langle n | \hat{\rho}_{I,12} D(t)^2 | n \rangle . \quad \text{(S44)}$$

IV. Calculate the density matrix elements up to the second order

We will proceed with the calculation of high harmonic emissions in the interaction frame. Following the same procedure of diagonalization and Dyson expansion up to the second order [5], we obtain $\hat{\rho}_I \approx \hat{\rho}_I^{(0)} + \hat{\rho}_I^{(1)} + \hat{\rho}_I^{(2)}$ as the following

$$\hat{\rho}_I^{(0)} = V \exp[i\sigma_z S(t)] \hat{\rho}_I(t=0) \exp[-i\sigma_z S(t)] V^+ \quad \text{(S45)}$$

$$\hat{\rho}_I^{(1)} = V \exp[i\sigma_z S(t)] \left[\int_{-\infty}^t \mathcal{W}(t_1) dt_1 \hat{\rho}_I(t=0) + \hat{\rho}_I(t=0) \int_{-\infty}^t \mathcal{W}^\dagger(t_1) dt_1 \right] \exp[-i\sigma_z S(t)] V^+ \quad \text{(S46)}$$

$$\hat{\rho}_I^{(2)} = V \exp[i\sigma_z S(t)] \left\{ \int_{t'}^t dt_1 \left[\mathcal{W}(t_1) \int_{t'}^{t_1} dt_2 \mathcal{W}(t_2) \right] \hat{\rho}_I(t=0) + \hat{\rho}_I(t=0) \int_{t'}^t dt_1 \left[\mathcal{W}^\dagger(t_1) \int_{t'}^{t_1} dt_2 \mathcal{W}^\dagger(t_2) \right] \right.$$
$$\left. + \int_{-\infty}^t \mathcal{W}(t_1) dt_1 \tilde{\rho}_0' \int_{-\infty}^t \mathcal{W}^\dagger(t_1) dt_1 \right\} \exp[-i\sigma_z S(t)] V^+. \quad \text{(S47)}$$

In particular, by denoting the energy difference of the two-level system as $\mathcal{E}$, we have used

$$V = \begin{bmatrix} V_1 & -V_2 {D^\dagger(t)}^2 \\ V_2 D(t)^2 & V_1 \end{bmatrix}, \quad V_1 = \sqrt{\frac{\mathcal{E} + \mathcal{E}_s}{2\mathcal{E}_s}}, \quad V_2 = \frac{-\hbar\Omega}{\sqrt{2\mathcal{E}_s(\mathcal{E} + \mathcal{E}_s)}}, \quad \text{(S48)}$$

$$S(t) = \int_{-\infty}^t \mathcal{E}_s(\tau)/(2\hbar) d\tau, \quad \mathcal{E}_s = \sqrt{\mathcal{E}(t)^2 + \hbar^2 \Omega(t)^2} \quad \text{(S49)}$$

$$\mathcal{W} = \exp[-i\sigma_z S(t)] \left(\partial_t V^\dagger \right) V \exp[i\sigma_z S(t)]. \quad \text{(S50)}$$

We introduce the dot derivative $\partial_t O = \dot{O}$ and write the $\int_{-\infty}^t \mathcal{W}(\tau) d\tau$ as

$$\int_{-\infty}^t \mathcal{W}(\tau) d\tau = \begin{bmatrix} \int_{-\infty}^t \mathcal{W}_{11} d\tau & \int_{-\infty}^t \mathcal{W}_{12} d\tau \\ -\int_{-\infty}^t \mathcal{W}_{12}^\dagger d\tau & -\int_{-\infty}^t \mathcal{W}_{11} d\tau \end{bmatrix} - \mathbb{1} \int_{-\infty}^t d\tau \mathcal{F}(\tau) + \sigma_z \int_{-\infty}^t d\tau V_1(\tau) \dot{V}_1(\tau) + V_2^*(\tau) \dot{V}_2(\tau) \quad \text{(S51)}$$

$$\int_{-\infty}^t \mathcal{W}_{11}(t') dt' = \frac{1}{2} \int_{-\infty}^t \left[1 - \frac{\mathcal{E}(t')}{\mathcal{E}_s(t')} \right] \left\{ \sum_q \frac{i2g_q}{\hbar} \left[b_q^\dagger(t') + b_q(t') \right] \right\} dt' \quad \text{(S52)}$$

$$\int_{-\infty}^t dt' \mathcal{F}(t') = \int_{-\infty}^t \frac{1}{2} \sum_q \frac{i4g_q^2}{\hbar^2 \omega_q} \left[1 - \frac{\mathcal{E}(t')}{\mathcal{E}_s(t')} \right] dt' \quad \text{(S53)}$$

$$\int_{-\infty}^t \mathcal{W}_{12}(t') dt' = \int_{-\infty}^t \exp[-2iS(t')] f_1(t') {D^\dagger(t')}^2 dt' + f_2(t) {D^\dagger}^2(t), \quad \text{(S54)}$$

$$f_1(t) = -2V_2 \dot{V}_1 - \frac{i\Omega}{2}, \quad f_2(t) = -\frac{\hbar\Omega \exp[-2iS(t)]}{2\mathcal{E}_s}. \quad \text{(S55)}$$

Note that the last two non-operator terms at the end of Eq.(S51) do not contribute to ρ and nonlinear current because the non-operator terms cancel all in Eqs.(S46,S47). Equations(S45-S47) can be cast into the form

$$\hat{\rho}_I = \begin{bmatrix} \hat{\rho}_{I,11} & \hat{\rho}_{I,12} \\ \hat{\rho}_{I,21} & \hat{\rho}_{I,22} \end{bmatrix} = V \exp[i\sigma_z S(t)] \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \exp[-i\sigma_z S(t)] V^\dagger. \quad \text{(S56)}$$

By writing out the right-hand side of the above equation, we have where

$$\hat{\rho}_{I,11} = V_1^2 M_{11} - \exp[2iS(t))] M_{12} V_1 V_2^* D^2(t) - \exp[-2iS(t)] V_1 V_2 D^\dagger(t)^2 M_{21} + |V_2|^2 D^\dagger(t)^2 M_{22} D(t)^2 \quad \text{(S57)}$$

$$\hat{\rho}_{I,12} = \hat{\rho}_{I,21}^\dagger = V_1 V_2 M_{11} D^\dagger(t)^2 + \exp[2iS(t)] V_1^2 M_{12} - \exp[-2iS(t)] {V_2}^2 D^\dagger(t)^2 M_{21} D^\dagger(t)^2 - V_1 V_2 D^\dagger(t)^2 M_{22} \quad \text{(S58)}$$

$$\hat{\rho}_{I,22} = |V_2|^2 D(t)^2 M_{11} D^\dagger(t)^2 + \exp[2iS(t)] V_1 V_2^* D(t)^2 M_{12} + \exp[-2iS(t)] V_1 V_2 M_{21} D^\dagger(t)^2 + V_1^2 M_{22}. \quad \text{(S59)}$$

With the above equations and owing to the electron number conservation, we have

$$\mathrm{Tr_B}[\hat{\rho}_{I,11}+\hat{\rho}_{I,22}] = \sum_n \langle n|\,\hat{\rho}_{I,11}+\hat{\rho}_{I,22}\,|n\rangle = \sum_n \langle n|\,M_{11}+M_{22}\,|n\rangle = 1. \tag{S60}$$

Combining Eqs. (S57-S60) and Eq.(S44), we obtain the current $j = j_{\rm intra} + j_{\rm inter}$ as the following

$$\begin{aligned} j_{\rm intra} &= -e\left\{v_{11}\mathrm{Tr}[\hat{\rho}_{I,11}] + v_{22}\mathrm{Tr}[\hat{\rho}_{I,22}]\right\} = -e\left(V_1^2 v_{11}\mathrm{Tr}[M_{11}] + |V_2|^2 v_{11}\mathrm{Tr}[M_{22}] + |V_2|^2 v_{22}\mathrm{Tr}[M_{11}] + V_1^2 v_{22}\mathrm{Tr}[M_{22}]\right. \\ &\quad \left. +2(v_{22}-v_{11})\mathrm{Re}\left\{V_1V_2^*\exp\left[2iS(t)\right]\mathrm{Tr}[M_{12}D^2(t)]\right\}\right) \end{aligned} \tag{S61}$$

$$\begin{aligned} j_{\rm inter} &= -2e\mathrm{Re}\left\{v_{12}\mathrm{Tr_B}[D^\dagger(t)^2\hat{\rho}_{I,21}]\right\} = -e2\mathrm{Re}\left\{v_{21}\mathrm{Tr_B}[D(t)^2\hat{\rho}_{I,12}]\right\} \\ &= -2e\mathrm{Re}\left\{v_{21}V_1V_2\mathrm{Tr_B}[M_{11}-M_{22}] + v_{21}V_1^2\mathrm{Tr_B}[\exp\left[2iS(t)\right]M_{12}D^2(t)] - v_{21}V_2^2\mathrm{Tr_B}[\exp\left[-2iS(t)\right]D^\dagger(t)^2M_{21}]\right\} \end{aligned} \tag{S62}$$

A. Density matrix elements for different orders

Calculation of the 0 order current: From Eq. (S45), we obtain

$$\hat{\rho}_I^{(0)} = \begin{bmatrix} V_1^2\mathcal{B}, & V_1V_2\mathcal{B}D^\dagger(t)^2 \\ V_1V_2^*D(t)^2\mathcal{B}, & |V_2|^2D(t)^2\mathcal{B}D^\dagger(t)^2 \end{bmatrix}. \tag{S63}$$

By denoting the density matrix elements before and after tracing over the photon number states $|n\rangle$ with a hat $\hat{}$ on the variable, i.e. $\sum_n \langle n|\,\hat{\rho}^{(0)}_{I,ij}\,|n\rangle = \rho^{(0)}_{I,ij}$, Eq. (S63) suggests that

$$\sum_n \langle n|\,\hat{\rho}^{(0)}_{I,11}\,|n\rangle = \rho^{(0)}_{I,11} = V_1^2, \quad \rho^{(0)}_{I,22} = |V_2|^2 \tag{S64}$$

$$\rho^{(0)}_{I,11} + \rho^{(0)}_{I,22} = 1 \tag{S65}$$

$$\rho^{(0)}_{I,12} = \rho^{(0)*}_{I,21}. \tag{S66}$$

Combining Eqs.(S61,S62,S45) , we obtain the 0 order current as

$$j^{(0)} = -e\left[V_1^2v_{11} + |V_2|^2v_{22} + 2\mathrm{Re}(V_1V_2v_{21})\right]. \tag{S67}$$

It is important to notice that since the environment is normalized i.e., $\mathrm{Tr_B}[\mathcal{B}]=1$, both the thermal heat bath and the squeezed vacuum lead to the same expression of the 0th order current as in Eq.(S67).

Calculation of the 1st order current: From Eq. (S60), we know that $\rho_{I,11}+\rho_{I,22}=1$ owing to the electron number conservation and $\rho_{I,12}=\rho^*_{I,21}$ owing to hermicity. As a result with $\rho^{(0)}_{I,11}+\rho^{(0)}_{I,22}=1$, all the higher orders density matrix elements of $\rho^{(n)}_{I,ij}$, $n>0$ have the relations

$$\rho^{(n)}_{I,11} = -\rho^{(n)}_{I,22}, \quad \rho^{(n)}_{I,12} = \rho^{(n)*}_{I,21}. \tag{S68}$$

According to Eq.(S68), for the calculations of $\rho^{(1)}_{I,ij}$ and $\rho^{(2)}_{I,ij}$, we will only focus on the M_{11} and M_{12} elements as shown in Eq.(S56). Following the same process as [5], we obtain

$$M^{(1)}_{11} = [\int_{-\infty}^t \mathcal{W}_{11}(t')dt', \mathcal{B}], \quad M^{(1)}_{12} = -\mathcal{B}\int_{-\infty}^t \mathcal{W}_{12}(t')dt' \tag{S69}$$

Note that $\mathrm{Tr_B}[M^{(1)}_{11}]=0$. Using the above relations and Eqs.(S61,S62) we obtain

$$j^{(1)}_{\rm intra} = 2e(v_{22}-v_{11})\mathrm{Re}\left\{V_1V_2^*\exp\left[2iS(t)\right]\mathrm{Tr_B}[\mathcal{B}\int_{-\infty}^t \mathcal{W}_{12}(t')dt'D^2(t)]\right\} \tag{S70}$$

$$j^{(1)}_{\rm inter} = -2e\mathrm{Re}\left\{v_{21}V_1^2\mathrm{Tr_B}[\exp\left[2iS(t)\right]\mathcal{B}\int_{-\infty}^t \mathcal{W}_{12}(t')dt'D^2(t)] - v_{21}V_2^2\mathrm{Tr_B}[\exp\left[-2iS(t)\right]D^\dagger(t)^2\int_{-\infty}^t \mathcal{W}^\dagger_{12}(t')dt'\mathcal{B}]\right\} \tag{S71}$$

<u>**Calculation of the 2nd order current:**</u> With $M_{11}^{(2)} = -M_{22}^{(2)}$ and Eqs.(S62,S61), we obtain

$$j^{(2)}_{\text{intra}} = -e(v_{11} - v_{22})\left((V_1^2 - |V_2|^2)\text{Tr}_\text{B}[M_{11}^{(2)}] - 2V_1V_2\text{Re}\left\{\exp\left[2iS(t)\right]\text{Tr}_\text{B}[M_{12}^{(2)}D^2(t)]\right\}\right) \tag{S72}$$

$$j^{(2)}_{\text{inter}} = -2e\text{Re}\left\{2v_{21}V_1V_2\text{Tr}_\text{B}[M_{11}^{(2)}] + v_{21}V_1^2\text{Tr}_\text{B}[\exp\left[2iS(t)\right]M_{12}^{(2)}D^2(t)] - v_{21}V_2^2\text{Tr}_\text{B}[\exp\left[-2iS(t)\right]{D^\dagger}^2(t)M_{21}^{(2)}]\right\}, \tag{S73}$$

where

$$\text{Tr}_\text{B}[M_{11}^{(2)}] = -\text{Tr}_\text{B}[M_{22}^{(2)}] = -\text{Tr}_\text{B}\left[\int_{-\infty}^t dt'\mathcal{W}^\dagger{}_{12}(t')\mathcal{B}\int_{-\infty}^t dt'\mathcal{W}_{12}(t')\right] \tag{S74}$$

$$= \int_{-\infty}^t\int_{-\infty}^t \exp\left[2iS(t_1,t_2)\right]f_1^*(t_1)f_1(t_2)\mathcal{C}_1(t_1,t_2)dt_1dt_2 + 2\text{Re}\left\{\int_{-\infty}^t \exp\left[2iS(t_1)\right]f_1^*(t_1)f_2(t)\mathcal{C}_1(t_1,t)dt_1\right\} + |f_2(t)|^2 \tag{S75}$$

$$M_{12}^{(2)} = \int_{-\infty}^t\int_{-\infty}^{t_1}\mathcal{B}\mathcal{W}_{11}(t_1)\mathcal{W}_{12}(t_2)dt_2dt_1 - \int_{-\infty}^t\int_{-\infty}^{t_1}\mathcal{B}\mathcal{W}_{12}(t_1)\mathcal{W}_{11}(t_2)dt_2dt_1 - \int_{-\infty}^t\mathcal{W}_{11}(t_1)dt_1\mathcal{B}\int_{-\infty}^t\mathcal{W}_{12}(t_1)dt_1. \tag{S76}$$

V. Current using the thermal heat bath

Substituting Eq.(S39) into $\mathcal{B}$, we obtain

$$\text{Tr}_\text{B}[BD(t)^2] = \exp\left[-\frac{1}{2}\sum_q\left|\frac{2g_q}{\hbar\omega_q}\right|^2\coth(\hbar\beta\omega_q/2)\right],$$

where $\beta = 1/(K_bT)$, K_b is the Boltzmann constant and T is the temperature. As a result, we have

$$\mathcal{R}_T(t,t') = \text{Tr}_\text{B}[BD^\dagger(t')^2D(t)^2] = \exp\left\{\sum_q\left|\frac{2g_q}{\hbar\omega_q}\right|^2\left[i\sin\left[\omega_q(t-t')\right] - \{1-\cos\left[\omega_q(t-t')\right]\}\coth\left(\hbar\beta\omega_q/2\right)\right]\right\} \tag{S77}$$

$$\mathcal{C}_T(t) = \text{Tr}_\text{B}\left\{\exp\left[2iS(t)\right]\mathcal{B}\int_{-\infty}^t\mathcal{W}_{12}(t')dt'D^2(t)\right\} = \int_{-\infty}^t dt'\exp\left[2iS(t,t')\right]f_1(t')\mathcal{R}_T(t,t') - \frac{\hbar\Omega(t)}{2\mathcal{E}_s(t)}. \tag{S78}$$

Using Eq.(S77), Eqs.(S70, S71) reduce to

$$j^{(1)}_{\text{intra}} = 2e(v_{22} - v_{11})\text{Re}\left[V_1V_2^*\mathcal{C}_\mathcal{T}(t)\right] \tag{S79}$$

$$j^{(1)}_{\text{inter}} = -2e\text{Re}\left\{v_{21}V_1^2\mathcal{C}_\mathcal{T}(t) - v_{21}V_2^2\mathcal{C}_\mathcal{T}^*(t)\right\}. \tag{S80}$$

In order to proceed with $j^{(2)}$, let's first look at the term $\text{Tr}_\text{B}[M_{12}^{(2)}D^2(t)]$. Take the first term in Eq.(S76) as an example, we have

$$\text{Tr}_\text{B}[\int_{-\infty}^t\int_{-\infty}^{t_1}\mathcal{B}\mathcal{W}_{11}(t_1)\mathcal{W}_{12}(t_2)D(t)^2dt_2dt_1] = \text{Tr}_\text{B}[\int_{-\infty}^t\int_{-\infty}^{t_1}\mathcal{B}\mathcal{W}_{11}(t_1)\exp\left[-2iS(t_2)\right]f_1(t_2)D^\dagger(t_2)^2D(t)^2dt_2dt_1]$$

$$= \frac{1}{2}\int_{-\infty}^t\int_{-\infty}^{t_1}\int_{-\infty}^{t_1}\left[1 - \frac{\mathcal{E}(t')}{\mathcal{E}_s(t')}\right]\exp\left[-2iS(t_2)\right]f_1(t_2)\sum_q\frac{i2g_q}{\hbar}\text{Tr}_\text{B}\left\{\mathcal{B}\left[b_q^\dagger(t') + b_q(t')\right]D^\dagger(t_2)^2D(t)^2\right\}dt_2dt_1dt'. \tag{S81}$$

To proceed, let's first define

$$D^\dagger(t)^2 = \exp\left[\sum_q\frac{2g_q^*}{\hbar\omega_q}\exp\left(i\omega_qt\right)b_q^\dagger - \frac{2g_q}{\hbar\omega_q}\exp\left(-i\omega_qt\right)b_q\right] = \exp\left[\sum_q\alpha_q(t)b_q^\dagger - \alpha_q(t)^*b_q\right], \tag{S82}$$

$$D^\dagger(t_2)^2D(t)^2 = \exp\left\{\sum_q\beta_qb_q^\dagger - \beta_q^*b_q - \frac{1}{2}[\alpha_q(t_2)\alpha_q(t)^* - \alpha_q(t_2)^*\alpha_q(t)]\right\} \tag{S83}$$

where $\alpha_a(t) = \frac{2g_q}{\hbar\omega_q}\exp(i\omega_q t)$, $\beta_q = \alpha_q(t_2) - \alpha_q(t)$. As a result, using the relation [11]

$$\partial_t \exp(\hat{O}) = \left\{\partial_t \hat{O} + \frac{1}{2}[\hat{O}, \partial_t \hat{O}]\right\}\exp(\hat{O}),$$

if the commutator [] gives a number, we obtain

$$\frac{\partial \exp\left(\sum_q \beta_q b_q^\dagger - \beta_q^* b_q\right)}{\partial \beta_q} = (b_q^\dagger - \beta_q^*/2)\exp\left(\sum_q \beta_q b_q^\dagger - \beta_q^* b_q\right). \tag{S84}$$

The trace part in Eq.(S81) reduces to

$$\begin{aligned}\mathrm{Tr_B}\left\{\mathcal{B} b_q^\dagger D^\dagger(t_2)^2 D(t)^2\right\} &= \exp\left\{\sum_q \left|\frac{2g_q}{\hbar\omega_q}\right|^2 i\sin[\omega_q(t-t_2)]\right\}\\ &\times\left\{\partial\exp\left[-\frac{|\beta_q|^2}{2}\coth(\hbar\beta\omega_q/2)\right]/\partial\beta_q + \frac{\beta_q^*}{2}\exp\left[-\frac{|\beta_q|^2}{2}\coth(\hbar\beta\omega_q/2)\right]\right\}.\end{aligned} \tag{S85}$$

Here, we focus on the leading contributions to the harmonic emissions. By observing Eqs.(S85, S81), knowing that Eq.(S81) i.e. $\mathrm{Tr_B}[M_{12}^{(2)} D^2(t)]$ is proportional to [5]

$$\left[1 - \frac{\mathcal{E}}{\mathcal{E}_s}\right] f_1 \sum_q \left|\frac{2g_q}{\hbar\omega_q}\right|^2 \delta(\omega - \omega_q) \ll f_1^* f_1 \tag{S86}$$

where the right-hand-side is proportional to $\mathrm{Tr_B}[M_{11}^{(2)}]$. As a result, for the second-order current $j^{(2)}$, we neglect all the $\mathrm{Tr_B}[M_{12}^{(2)} D^2(t)]$ related terms. In particular, we have chosen the Ohmic spectral density

$$\sum_q \left|\frac{2g_q}{\hbar\omega_q}\right|^2 \delta(\omega - \omega_q) = \int d\omega \frac{j_0\omega}{\omega_c^2}\exp(-|\omega|/\omega_c), \quad G_T(\omega) = \frac{j_o\omega}{\omega_c^2}\exp(-|\omega|/\omega_c). \tag{S87}$$

Here, the j_o and ω_c represent the coupling strength and the cut-off frequency. In particular, we have chosen room temperature $T = 300$ K and strong coupling coefficient $j_o = 2$ with $\omega_c = 2.1\omega_0$ for all the calculations in the main text. To summarize, since we know that $\hbar\Omega \ll \mathcal{E}_s$, i.e. $1 - \mathcal{E}(t')/\mathcal{E}_s(t') \ll 1$ owing to under-resonant conditions, we have the following approximations

$$f_1(t) \approx -i\Omega(t)/2, \tag{S88}$$

$$\mathcal{C}_T \approx \int_{-\infty}^{t} \frac{-i\Omega(t_1)}{2}\exp[2iS(t,t_1)]\mathcal{R}_T(t,t_1)dt_1 - \frac{\hbar\Omega(t)}{2\mathcal{E}_s(t)}, \tag{S89}$$

$$\text{where, } \mathcal{R}_T(t,t_1) = \exp\left\{\int_{-\infty}^{\infty} G_T(\omega)\left[i\sin[\omega(t-t_1)] - \{1-\cos[\omega(t-t_1)]\}\coth(\hbar\beta\omega/2)\right]d\omega\right\}. \tag{S90}$$

As a result, we obtain

$$j_{\text{intra}}^{(0)} = -e[v_{11}V_1^2 + |V_2|^2 v_{22}], \quad j_{\text{inter}}^{(0)} = -2e\mathrm{Re}(V_1 V_2 v_{21}) \tag{S91}$$

$$j_{\text{intra}}^{(1)} = 2e(v_{22} - v_{11})\mathrm{Re}[V_1 V_2^* \mathcal{C}_T(t)], \quad j_{\text{inter}}^{(1)} = -2e\mathrm{Re}\left[v_{21}V_1^2\mathcal{C}_T(t) - v_{21}V_2^2\mathcal{C}_T^*(t)\right] \tag{S92}$$

$$\begin{aligned}j_{\text{intra}}^{(2)} &= \frac{e}{4}(v_{22} - v_{11})(V_1^2 - |V_2|^2)\int_{-\infty}^{t}\int_{-\infty}^{t}\exp[2iS(t_1,t_2)]\Omega^*(t_1)\Omega(t_2)\mathcal{R}_T(t_1,t_2)dt_1dt_2\\ &= \frac{e}{2}(v_{22} - v_{11})(V_1^2 - |V_2|^2)\int_{-\infty}^{t}\int_{-\infty}^{t_1}\exp[2iS(t_1,t_2)]\Omega^*(t_1)\Omega(t_2)\mathcal{R}_T(t_1,t_2)dt_1dt_2\end{aligned} \tag{S93}$$

$$j_{\text{inter}}^{(2)} = -e\mathrm{Re}\left\{v_{21}V_1V_2\int_{-\infty}^{t}\int_{-\infty}^{t}\exp[2iS(t_1,t_2)]\Omega^*(t_1)\Omega(t_2)\mathcal{R}_T(t_1,t_2)dt_1dt_2\right\} \tag{S94}$$

VI. Current using the squeezed vacuum

For the environment as the squeezed vacuum, we have $\mathcal{B} = D^\dagger S(\xi)\left|0\right\rangle\left\langle 0\right| S^{-1}(\xi) D$ as shown in Eq.(S38). We now focus on the first order of current in Eqs(S70,S71). By omitting the terms with $\hbar\Omega/\mathcal{E}_s$, the results reduce to obtaining

$$\mathrm{Tr_B}\left\{D^\dagger S(\xi)\left|0\right\rangle\left\langle 0\right| S^\dagger(\xi) D D^\dagger(t')^2 D^2(t)\right\} = \left\langle 0\right| S^\dagger(\xi) D D^\dagger(t')^2 D^2(t) D^\dagger S(\xi)\left|0\right\rangle. \tag{S95}$$

One can obtain after some misery that

$$\begin{aligned}
&\left\langle 0\right|\hat{S}^\dagger(\xi)\exp\left[\sum_q \beta_q b_q^\dagger - \beta_q^* b_q\right]\hat{S}(\xi)\left|0\right\rangle \\
&= \exp\left\{-\frac{1}{2}\sum_{q,j,q'}[\beta_q(\cosh\widetilde{\boldsymbol{r}})_{q,j} + \beta_q^*(e^{i\boldsymbol{\theta}}\sinh\boldsymbol{r})_{qj}][\beta_{q'}^*(\cosh\boldsymbol{r})_{q',j} + \beta_{q'}(\sinh\widetilde{\boldsymbol{r}}e^{-i\widetilde{\boldsymbol{\theta}}})_{q',j}]\right\}, \qquad \text{(S96)}\\
&\approx \exp\left\{-\sum_q \frac{|\beta_q|^2}{2}[\cosh^2 r_q + \sinh^2 r_q + 2\cosh r_q \sinh r_q \cos(2\theta_q - \theta)]\right\}. \qquad \text{(S97)}
\end{aligned}$$

The last step shown in Eq.(S97) is an approximation and is valid when the brightly squeezed vacuum pulse is very weak. Here, we want to associate an effective squeezing parameter with each frequency. Using the relation

$$D^\dagger(t')^2 D^2(t) = \exp\left(\sum_q i\left|\frac{2g_q}{\hbar\omega_q}\right|^2\{\sin\left[\omega_q(t-t')\right]\}\right)\exp\left(\sum_q \beta_q b_q^\dagger - \beta_q^* b_q\right),$$

where $\beta_q = \frac{2g_q^*}{\hbar\omega_q}[\exp(i\omega_q t') - \exp(i\omega_q t)]$, and that

$$\exp\left[-\sum_q (A_q b_q^\dagger - A_q^* b_q)\right]\exp\left[\sum_q B_q b_q^\dagger - B_q^* b_q\right]\exp\left[\sum_q A_q b_q^\dagger - A_q^* b_q\right] = \exp\left[\sum_q B_q b_q^\dagger - B_q^* b_q\right]\exp\left[\sum_q B_q A_q^* - B_q^* A_q\right], \tag{S98}$$

we can obtain

$$D D^\dagger(t')^2 D^2(t) D^\dagger = \exp\left(\sum_q i\left|\frac{2g_q}{\hbar\omega_q}\right|^2\{\sin\left[\omega_q(t-t')\right] + \sin(\omega_q t') - \sin(\omega_q t)\}\right)\exp\left(\sum_q \beta_q b_q^\dagger - \beta_q^* b_q\right). \tag{S99}$$

Combining Eqs(S99,S97), Eq.(S95) reduces to

$$\begin{aligned}
\mathcal{R}_S(t,t') = \left\langle 0\right| S^\dagger(\xi) D D^\dagger(t')^2 D^2(t) D^\dagger S(\xi)\left|0\right\rangle &= \exp\left(\sum_q i\left|\frac{2g_q}{\hbar\omega_q}\right|^2\{\sin\left[\omega_q(t-t')\right] + \sin(\omega_q t') - \sin(\omega_q t)\}\right)\\
\times\exp\left[\sum_q -\left|\frac{2g_q}{\hbar\omega_q}\right|^2\{1-\cos\left[\omega_q(t-t')\right]\}\{\cosh 2r_q + \cos\left[2\theta_q(t,t')-\theta\right]\sinh 2r_q\}\right]&, \qquad \text{(S100)}
\end{aligned}$$

where $\tan\left[\theta_q(t,t')\right] = [\sin(t'\omega_q) - \sin(t\omega_q)]/[\cos(t'\omega_q) - \cos(t\omega_q)]$. Note that the real part of Eq.(S100), i.e. the second exponential term, is always a decay. In other words, the value inside this exponential function is always negative. Remember that $M_{11}^{(1)} = 0$, as a result, by defining

$$\mathcal{C}_S \approx \int_{-\infty}^{t}\frac{-i\Omega(t_1)}{2}\exp\left[2iS(t,t_1)\right]\mathcal{R}_S(t,t_1)dt_1 - \frac{\hbar\Omega(t)}{2\mathcal{E}_s(t)}, \tag{S101}$$

we obtain the first-order current of the squeezed vacuum environment as

$$j_{\text{intra}}^{(1)} = 2e(v_{22} - v_{11})\mathrm{Re}\left[V_1 V_2^* \mathcal{C}_S(t)\right] \tag{S102}$$

$$j_{\text{inter}}^{(1)} = -2e\mathrm{Re}\left[v_{21}V_1^2\mathcal{C}_S(t) - v_{21}V_2^2\mathcal{C}_S^*(t)\right] \tag{S103}$$

Following the same steps for the second order current as Section V, we need to obtain

$$\mathrm{Tr_B}\left\{\mathcal{B}b_q^\dagger D^\dagger(t_2)^2 D(t)^2\right\} = \exp\left(\sum_q i\left|\frac{2g_q}{\hbar\omega_q}\right|^2\left\{\sin\left[\omega_q(t-t')\right] + \sin\left(\omega_q t'\right) - \sin\left(\omega_q t\right)\right\}\right)$$
$$\times\left\{\partial\exp\left(-\frac{1}{2}\sum_q\left|\beta_q\cosh r + \beta_q^* e^{i\theta}\sinh r\right|^2\right)/\partial\beta_q + \frac{\beta_q^*}{2}\exp\left(-\frac{1}{2}\sum_q\left|\beta_q\cosh r + \beta_q^* e^{i\theta}\sinh r\right|^2\right)\right\}. \tag{S104}$$

To proceed with the calculation and throw away small terms, we want to use the same argument as in Section V Eq.(S86).

A. Define the relation of spectrum and squeezing parameters

Typically, the squeezed light is generated via optical parametric amplification [12]. By writing the single-mode/frequency electric field operator as

$$\hat{E}_q = E_{0,q}\left[a_q^\dagger\exp(i\chi_q) + a_q\exp(-i\chi_q\right] \tag{S105}$$

where the subscript q represents a given frequency, $E_{0,q} = \sqrt{\hbar\omega_q/(2V\epsilon_0)}$ with V as the quantized volum [6] ,$a_q^\dagger, a_q$ are creation and annihilation operators, $\chi_q = \pi/2 + \omega_q t - k_q x$, and x is the position. The fluctuation for the squeezed vacuum is defined as

$$\Delta E_q^2 = \langle\xi|\,\hat{E}_q^2\,|\xi\rangle - \langle\xi|\,\hat{E}_q\,|\xi\rangle^2 = \langle\xi|\,\hat{E}_q^2\,|\xi\rangle = E_{0,q}^2\left[\cosh 2r_q - \sinh 2r_q\cos\left(2\chi_q - \theta\right)\right]. \tag{S106}$$

It is essential to notice that the energy and amplitude of the pulse are contained in the state $|\xi\rangle$, not in the electric field operator itself. The squeezing parameter is defined as $r_q\exp(i\theta)$, where r_q is a purely real number. In the following, we assume that all the squeezing phases are identical and $\theta = \pi/2$. Besides, the photon number of a given mode q is defined as

$$\langle\xi|\,\hat{n}_q\,|\xi\rangle = \langle\xi|\,a_q^\dagger a_q\,|\xi\rangle = \sinh(r_q)^2. \tag{S107}$$

We know that a multi-mode squeezed vacuum spectrum can be defined as

$$E(\omega) = E_{\omega,0}\exp\left[\frac{-(\omega-\omega_0)^2\tau^2}{4}\right], \tag{S108}$$

where $E_{\omega,0} = E_0\tau/\sqrt{2}$ in the unit of [V s/m]. We have

$$\int\frac{A}{2}c\epsilon_0|E(\omega)|^2 d\omega = \sum_q\hbar\omega_q\,\langle\xi|\,n_q\,|\xi\rangle\,, \tag{S109}$$

where a given squeezed vacuum beam size is A, and τ is the pulse duration. In the main text we have chosen $A = \pi(4.5\times10^{-4})^2$ [m^2] and $\tau = 40$ fs. By defining the pulse energy $U = Ac\epsilon_0 E_0^2\tau\sqrt{\pi/8} \approx 40\,\mathrm{nJ}$ and combining Eqs.(S107) and (S109), we obtain

$$\langle\xi|\,n_q\,|\xi\rangle = \frac{Ac\epsilon_0|E_{\omega,0}|^2}{2\hbar\omega_q}\exp\left[\frac{-(\omega_q-\omega_0)^2\tau^2}{2}\right]d\omega_q = \frac{U\tau}{\sqrt{2\pi}\hbar\omega_q}\exp\left[\frac{-(\omega_q-\omega_0)^2\tau^2}{2}\right]d\omega_q, \tag{S110}$$

$$\cosh\left(2r_q\right) = 1 + 2\,\langle\xi|\,n_q\,|\xi\rangle\,,\quad \sinh\left(2r_q\right) = \sqrt{\cosh\left(2r_q\right)^2 - 1} \tag{S111}$$

where $d\omega_q$ is the resolution window of a given frequency. With the above equations, we make a connection between the fluctuation and the energy of a given frequency. An illustration of summing over different modes q and the resulting electric field is shown in Fig.S4.

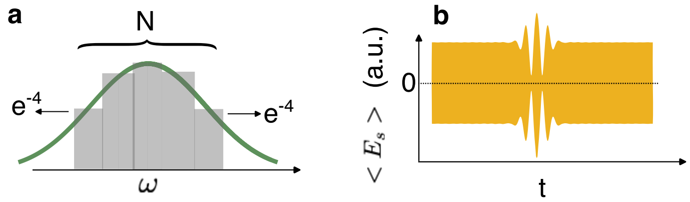

FIG. S4. Panel **a** illustrates the selection of frequency elements in a continuous spectrum. We have chosen evenly distributed 151 frequency mesh bins within the spectral range $[\omega_0\text{-}4/\tau,\omega_0\text{+}4/\tau]$ corresponding to the $\exp(-4)$. This frequency range is wide enough to cover most of the pulse. Panel **b** presents the fluctuation of the resulting electric field.

B. Define $\left|\frac{g_q}{\hbar\omega_q}\right|^2$

In order to obtain the coupling strength between the environment and the two-band system, we need to first dive into where this g_q parameter stems from. Here, we need to refresh our brains again and remind ourselves of the semiconductor Bloch wave equation in the co-moving frame as [13, 14]

$$\frac{\partial\rho_{nm}(K,t')}{\partial t'} = -i\omega_{nm}(K'_t)\rho_{nm}(K,t') + i\left[\sum_{l\neq n}\Omega_{nl}(K_{t'},t')\rho_{lm}(K,t') - \sum_{l\neq m}\Omega_{lm}(K_{t'},t')\rho_{nl}(K,t')\right]. \tag{S112}$$

For a two-band system, the above equation can be cast into the form

$$i\hbar\frac{\partial\rho}{\partial t'} = [H(K'_t,t'),\rho(K,t')], \tag{S113}$$

where

$$H = \frac{1}{2}\begin{bmatrix} -\hbar\omega_{21}(K'_t,t') & \hbar\Omega_{12}(K'_t,t') \\ \hbar\Omega_{21}(K'_t,t') & \hbar\omega_{21}(K'_t,t') \end{bmatrix} = -\frac{\hbar\omega_{21}(K'_t,t')}{2}\sigma_z + \frac{1}{2}\hbar\Omega(K'_t,t')\sigma_x. \tag{S114}$$

Equation (S114) is exactly our electron part i.e. the spin-boson model without the interaction and the boson environment. Since we know that $K'_t = K + |e|A(t')/\hbar$, we now consider $A(t') = A_c(t') + A_q(t')$, where the subscripts c and q represents "classical" and "quantum" respectively. In Eq.(S14), since the interaction part (last term) contains only the $a_j^\dagger a_j$, the environment only causes intra-band dephasing, and does not cause direct excitations. Thus, we only expand the σ_z related terms to the quantum vector potential with the following

$$\omega_{21}[K + \frac{|e|}{\hbar}(A_c(t') + A_q(t'))] \approx \omega_{21}[K + \frac{|e|}{\hbar}A_c(t')] + \frac{|e|}{\hbar}A_q(t')\nabla_K\omega_{21} = \omega_{21}[K + \frac{|e|}{\hbar}A_c(t')] + \frac{|e|}{\hbar}(v_{22} - v_{11})A_q(t') \tag{S115}$$

$$\Omega_{12}[K + \frac{|e|}{\hbar}(A_c(t') + A_q(t'))] \approx \Omega[K + \frac{|e|}{\hbar}A_c(t')], \tag{S116}$$

where $v_{ii} = \partial\mathcal{E}_i/(\hbar\partial K)$. With the knowledge $A_q = -\int E_q(\tau)d\tau$, combining with Eq.(S105), we obtain

$$A_q = \frac{-E_q}{i\omega_q}[a_q^\dagger\exp(i\chi_q) - a_q\exp(-i\chi_q)], \tag{S117}$$

where E_q is the electric field strength at mode q. Substituting Eqs(S117,S115) back into Eq(S114) and sum over all the q modes, we obtain

$$H = -\frac{\hbar\omega_{21}(K'_t,t')}{2}\sigma_z + \frac{1}{2}\hbar\Omega(K'_t,t')\sigma_x + \sigma_z\sum_q\frac{|e|(v_{22} - v_{11})E_{0,q}}{i2\omega_q}[a_q^\dagger\exp(i\chi_q) - a_q\exp(-i\chi_q)]. \tag{S118}$$

Comparing the last term from the above equation with the last term in Eq.(S14) [5], we immediately see that the coupling strength

$$g_q = \frac{|e|(v_{22} - v_{11})E_{0,q}}{2i\omega_q}\exp(i\chi_q). \tag{S119}$$

From Eq.(S119) we obtain

$$\sum_q \left|\frac{g_q}{\hbar\omega_q}\right|^2 = \frac{V}{(2\pi)^3}\int\left|\frac{g_q}{\hbar\omega_q}\right|^2 dk^3 = \int_0^\infty G_S(\omega)d\omega, \quad G_S(\omega) = \frac{e^2(v_{22}-v_{11})^2}{16\pi^2c^3\hbar\omega\epsilon_0}. \tag{S120}$$

To summarize, the current caused by a squeezed vacuum is

$$j^{(0)}_{\text{intra}} = -e[v_{11}V_1^2 + |V_2|^2 v_{22}], \quad j^{(0)}_{\text{inter}} = -2e\text{Re}(V_1V_2v_{21}) \tag{S121}$$

$$j^{(1)}_{\text{intra}} = 2e(v_{22}-v_{11})\text{Re}\left[V_1V_2^*\mathcal{C}_S(t)\right], \quad j^{(1)}_{\text{inter}} = -2e\text{Re}\left[v_{21}V_1^2\mathcal{C}_S(t) - v_{21}V_2^2\mathcal{C}_S^*(t)\right] \tag{S122}$$

$$j^{(2)}_{\text{intra}} = \frac{e}{4}(v_{22}-v_{11})(V_1^2-|V_2|^2)\int_{-\infty}^t\int_{-\infty}^t \exp\left[2iS(t_1,t_2)\right]\Omega^*(t_1)\Omega(t_2)\mathcal{R}_S(t_1,t_2)dt_1dt_2$$

$$= \frac{e}{2}(v_{22}-v_{11})(V_1^2-|V_2|^2)\int_{-\infty}^t\int_{-\infty}^{t_1} \exp\left[2iS(t_1,t_2)\right]\Omega^*(t_1)\Omega(t_2)\mathcal{R}_S(t_1,t_2)dt_1dt_2 \tag{S123}$$

$$j^{(2)}_{\text{inter}} = -e\text{Re}\left\{v_{21}V_1V_2\int_{-\infty}^t\int_{-\infty}^t \exp\left[2iS(t_1,t_2)\right]\Omega^*(t_1)\Omega(t_2)\mathcal{R}_S(t_1,t_2)dt_1dt_2\right\} \tag{S124}$$

where

$$V_1 = \sqrt{\frac{\mathcal{E}+\mathcal{E}_s}{2\mathcal{E}_s}}, \quad V_2 = \frac{-\hbar\Omega}{\sqrt{2\mathcal{E}_s(\mathcal{E}+\mathcal{E}_s)}}, \tag{S125}$$

$$S(t) = \int_{-\infty}^t \mathcal{E}_s(\tau)/(2\hbar)d\tau, \quad \mathcal{E}_s = \sqrt{\mathcal{E}(t)^2+\hbar^2\Omega(t)^2} \tag{S126}$$

$$\mathcal{R}_S(t,t_1) = \exp\left(\int_0^\infty iG_{\mathcal{S}}(\omega)\left\{\sin\left[\omega(t-t_1)\right] + \sin\left(\omega t_1\right) - \sin\left(\omega t\right)\right\}d\omega\right)$$

$$\times\exp\left[-\int_0^\infty G_{\mathcal{S}}(\omega)\left\{1-\cos\left[\omega(t-t_1)\right]\right\}\left\{\cosh[2r(\omega)] + \cos\left[2\theta_q(t,t_1)-\theta\right]\sinh[2r(\omega)]\right\}d\omega\right]$$

$$\approx\exp\left[-\int_0^\infty G_{\mathcal{S}}(\omega)\left\{1-\cos\left[\omega(t-t_1)\right]\right\}\left\{\cosh[2r(\omega)] + \cos\left[2\theta_q(t,t_1)-\theta\right]\sinh[2r(\omega)]\right\}d\omega\right] \tag{S127}$$

$$\mathcal{C}_S(t) \approx \int_{-\infty}^t \frac{-i\Omega(t_1)}{2}\exp\left[2iS(t,t_1)\right]\mathcal{R}_S(t,t_1)dt_1 - \frac{\hbar\Omega(t)}{2\mathcal{E}_s(t)} \tag{S128}$$

$$\tan\left[\theta_q(t,t_1)\right] = \frac{\sin(t_1\omega)-\sin(t\omega)}{\cos(t_1\omega)-\cos(t\omega)}. \tag{S129}$$

Equation (S127) can be considered purely real because the cosh() and sinh() are far larger than 1. As a result, the imaginary part can be ignored. Since θ corresponds to the squeezing phase, without loss of generality, it is set to $\pi/2$. We found that the value of θ does not influence the calculation results.

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